Modeling Estimates of the Effect of Acid Rain on Background Radiation Dose

by Stephen C. Sheppard* and Marsha I. Sheppard*

Acid rain causes accelerated mobilization of many materials in soils. Natural and anthropogenic radionuclides, especially ^{226}Ra and ^{137}Cs, are among these materials. Okamoto is apparently the only researcher to date who has attempted to quantify the effect of acid rain on the "background" radiation dose to man (1). He estimated an increase in dose by a factor of 1.3 following a decrease in soil pH of 1 unit. We reviewed literature that described the effects of changes in pH on mobility and plant uptake of Ra and Cs. Generally, a decrease in soil pH by 1 unit will increase mobility and plant uptake by factors of 2 to 7. Thus, Okamoto's dose estimate may be too low.

We applied several simulation models to confirm Okamoto's ideas, with most emphasis on an atmospherically driven soil model that predicts water and nuclide flow through a soil profile. We modeled a typical, acid-rain sensitive soil using meteorological data from Geraldton, Ontario. The results, within the range of effects on the soil expected from acidification, showed essentially direct proportionality between the mobility of the nuclides and dose. This supports some of the assumptions invoked by Okamoto. We conclude that a decrease in pH of 1 unit may increase the mobility of Ra and Cs by a factor of 2 or more. Our models predict that this will lead to similar increases in plant uptake and radiological dose to man. Although health effects following such a small increase in dose have not been statistically demonstrated, any increase in dose is probably undesirable.

Introduction

Radiation-Dose Consequences of Acid Rain

Acid rain has become one of the most prominent environmental problems caused by industrial activities. Its relatively slow and widespread detrimental effects have made it especially difficult to assess and abate. Much of the emphasis has been on aquatic environments where reduced buffering, thorough mixing, and high sensitivity of organisms have led to the most dramatic effects. However, terrestrial environments are also affected. Troedsson reported that although pH changes in forest soils were slight, marked losses of exchangeable Ca, Mg, and K had occurred, presumably due to acid rain (2). Troedsson's study was based on 10 years of data from 13,000 test plots in Sweden.

Direct effects on human health of acid rain have been examined. Drinking water can have increased concentrations of metals when acidified rainfall accelerates the leaching from soils and/or the corrosion of water supply systems, although health impacts have not been proven (3). Epidemiology studies in Canada

have also failed to statistically prove a relationship between acid rain and human health, although trends of detrimental effects were apparent (4). Few studies have considered radiological effects on man that may result from acid rain.

Reports from the Swedish nuclear industry have briefly alluded to the possibility of acid rain increasing the solubility and plant uptake of radionuclides (5,6). Okamoto has published the only attempt to quantify this impact (1). He estimated a dose increase of 14 μSv/a* when soil pH drops by 1 pH unit. Several comparable estimates for different scenarios are given in Table 1 (7). The concept is of particular interest in Ontario, Canada, where uranium mining activities

Table 1. Radiation dose to humans from technologically enhanced sources of natural radiation.[a]

Source	Dose, μSv/a
Household use of natural gas	20
Living near a coal-fired electrical generation plant	15
Living in a concrete house	30–200
Living on a uranium mine tailing	700

[a]From Stranden (7).

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*The unit Sv (Sievert) represents the dose equivalent and is the absorbed dose modified for the specific type of radiation.

and areas of naturally elevated radionuclides (8) overlap with areas sensitive to acid rain (9). Our work complements Okamoto's information on ^{226}Ra, a highly mobile anthropogenic radionuclide, and also includes ^{137}Cs, a fallout nuclide, present in Canadian soils.

Review of Okamoto's Analysis

Okamoto based his arguments on studies of the effect of acidic surface waters on leaching of ^{226}Ra from uranium mine slags. The studies were done with samples of two uranium ores and one uranium processing waste (10). Havlik et al. extracted 3 g of the pulverized samples with 30 mL of solution (10); the solution pHs were adjusted to range from 1 to 13. The acids HCl or H_3BO_4 were used to achieve pH values below 7.0. The time courses of the extractions were erratic (Fig. 1) and generally showed the highest extraction yield after 1 to 2 hr, followed by apparent resorption up to 5 hr. Havlik et al. (10) tabulated the maximum extraction fractions for each time course and Okamoto plotted these values, for the waste material, as shown in Figure 2. We independently estimated mean extraction fractions from the data of Havlik et al. to confirm Okamoto's procedures and conclusions.

Okamoto noted that the extraction fraction for Ra was approximately linear between pH 6 and 3. A drop in pH from 5 to 4 would change the fraction from 0.065 to 0.085 (0.02 per unit pH). This difference of 0.02 expressed as a percent of the extraction fraction at pH 5 (0.065) is approximately 30%. When we applied this approach to the estimated mean extraction fractions of Havlik et al. (10), the slope of 0.02 per unit pH was confirmed, but the extraction fraction at pH 5 was only about 0.035. Expressed as a percent, this is 57%. Thus, based on our analysis of the data of Havik et al. (10), a change in pH from 5 to 4 could increase the release of Ra in the range of 30 to 57%.
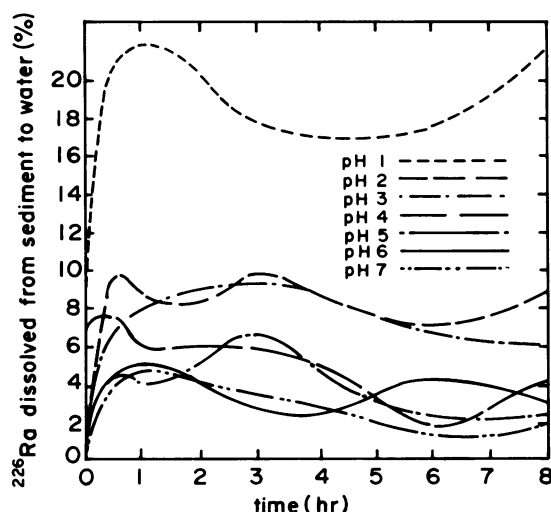

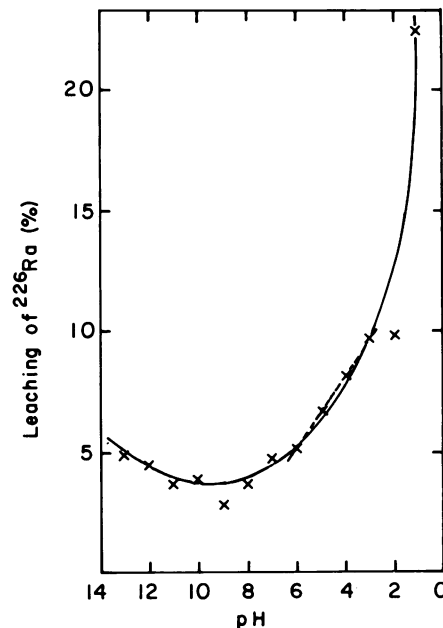FIGURE 1. Effect of pH on the ^{226}Ra liberation from sediments to acidified water. From Havlik et al. (10).

FIGURE 2. Relation between the ^{226}Ra leaching rate from uranium slag sediments and the pH. From Okamoto (1).

Okamoto assumed that the 30% increase in solubilization of Ra due to a pH drop from 5 to 4 applies to all sources of Ra that affect man. He further assumed that this leads to a 30% increase in dose due to Ra. Based on normal exposures from natural sources, this would result in an increase dose of 5.4 μSv/a due to Ra. He further assumed that doses from ^{238}U-^{234}U, ^{230}Th, and ^{232}Th are similarly affected by a change in pH and that this would add another 8.4 μSv/a. Okamoto recognized that several of his assumptions were tenuous.

The dose received by man from natural radionuclides corresponds to a near-equilibrium state in elemental cycling. Therefore, a shift in key elemental distributions, such as that between immobile (solid) and mobile (liquid) phases, could conceivably have a proportional effect throughout the cycle. Conversely, natural systems are replete with counter-balancing and feedback phenomena. Clearly, a more sophisticated approach is required to further assess radiation-dose enhancement by acid rain.

Literature Review

Here we review the relevant literature to establish a firm basis for simulating effects of acid rain on radiation exposure of man. Because there are a number of excellent reviews on the interaction of acid rain with soil, this subject will not be dealt with in detail (11-13).

Chemistry of Ra in the Environment

Reports by Sheppard (14) and Shoesmith (15) give considerable detail on the chemistry of Ra in the

environment. The key features of Ra chemistry are: a) It is generally present as an inorganic cation; b) its behavior is very similar to Ca^{2+}; c) its behavior is modified by the presence of Ca^{2+}, often in a competitive manner. When more Ca is present, the Ra is more effectively sorbed by soils and less effectively absorbed by plants; and d) it is more mobile at low soil pHs and is therefore more available for plant uptake.

Environmental concentrations of Ra are generally very low. Osburn (16) estimated concentrations of 10^{-6} μg Ra/g in granitic rocks and concentrations an order of magnitude higher are reported for ores, mine wastes (10), and mineral springs (14). Soils near Port Hope, Ontario, have up to 1.8×10^{-4} μg Ra/g (15). Usually less than 5% of the Ra is in the liquid phase, resulting in very low solution concentrations. As such, coprecipitation (probably with Ca) and adsorption/occlusion may limit the usefulness of usual water chemistry models to predict mobility.

Effects of Ca and pH on Ra. Okamoto (1) used data from Havlik et al. (10) as a basis for his dose estimates resulting from acid rain. Several other papers give similar relationships and may be preferable as they deal with soils, or plants in more natural settings, as opposed to mine wastes in a laboratory flask system.

Rusanova extracted native Ra from soils with buffer solutions varying in pH from 2.2 to 9.8 (17). A drop in pH from 5 to 4 approximately doubled the desorption of Ra. Even larger differences were found when comparisons were made of the relative solubility of Ra in four soils naturally varying in pH. The fraction of total Ra leached from surface soil samples using 1 mole/L KCl, and the corresponding pH values were 0.012% at pH 6.9; 0.006 and 0.009% at pH 6.6; and 0.066% at pH 5.9.

Several studies show a dramatic change in Ra mobility and/or plant uptake when pH changed in the range of 5 to 7 (18–21). Whereas Okamoto assumed a 1.3-fold increase in dissolved Ra following a drop in pH of 1 unit around pH 5 (1), other studies show 2- to 6-fold increases following a similar drop in pH. Thus, the conclusions of Okamoto may have been an underestimate.

Many of these studies deal with soils and solutions in isolation and may therefore not be directly relevant. In the real world, increased mobility of Ra will result in both increased plant uptake and increased leaching losses. There are no data in the literature to adequately examine pH effects on these two processes operating simultaneously. Thus, a modeling approach with emphasis on coupled water/nuclide flow processes is the most expedient means to assess these effects.

Chemistry and the Effect of pH on Cs in the Environment

The reactions of Cs in natural systems have been extensively reviewed (22,23). The key features of Cs

chemistry are: a) It is generally present as an inorganic cation; b) its behavior is similar to K^+ and NH_4^+ c); its behavior is modified in a competitive manner by the presence of K^+; d) it is subject to fixation, or nearly irreversible sorption, in clay materials, probably by means of entrapment between platelets of clay particles; and e) most ^{137}Cs is the product of atmospheric weapons testing; thus, its presence in soils is due to relatively recent atmospheric deposition. Since deposition, the levels of ^{137}Cs in surface soils have decreased due to leaching, erosion, and radioactive decay.

Benson et al. examined ^{137}Cs concentrations latitudinally across Canada (24). Vegetation contained an average of 185 Bq/m^{2*} of ^{137}Cs, with the highest concentrations at latitudes above 60°N.

Several papers have compared the mobility of Cs in different soils. Most emphasize clay soils, and therefore effects of pH or base saturation are confounded by effects of soil texture. Caplan et al. mentions a link between Cs uptake by plants and acid rain (25). They sampled 80 pine and spruce stands in Norway. One of their most distinct findings was that plant samples from southern Norway, in areas subjected to acid rain, had higher tissue concentrations of Cs than those from Central Norway. Since this is opposite to the general pattern of radioactive fallout, it may reflect a specific effect of acid rain. The largest effect was for pine needles, with about a sevenfold increase in Cs in the acid-rain affected area.

In general, the relationship between pH and Cs mobility is probably a simple cation exchange phenomenon (26). At low pH, the Cs on the exchange sites in the soil is replaced to a greater extent by hydrogen than at high pH (27,28). Thus, soil acidification will likely cause mobilization of Cs. Based on the information available, this is very difficult to quantify. Considering a simple cation exchange model, a 10-fold change in hydrogen ion concentration (1 pH unit) could displace 90% of the exchangeable Cs.

Analogy with Other Materials Solubilized by Acid Rain

It is simplistic to conclude that if pH controls mobility of an element in the lab, that this will result in differences in plant uptake in the field under acid rain conditions. Rechcigl and Sparks (12), in their recent review of the effects of acid rain on soils conclude that enhanced mobilization and uptake of harmful metals is still an open question, despite supporting laboratory data. The concept has received considerable attention (29–32), with particular emphasis on Hg, Al, Ni, Cd, Zn, Mn, As, and Se. Few studies have shown an effect of acid rain on metal uptake by plants in the field. Ulrich et al. noted Al toxicity on beech tree roots as a result of soil acidification (33).

In general, an element with increased mobility will

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*The unit Bq (Bequerel) represents the number of radioactive disintegrations per second.

be more susceptible to both leaching from the soil profile and plant uptake. The exact fate of the mobilized element becomes a function of the hydraulic regime in the soil. Thus, an assessment of the fate of radionuclides released by acid rain must pay careful attention to the water flow in the soil system.

Consequences of Increased Background Radiation Dose

The general principle guiding radiation protection is to reduce doses as much as reasonably possible (34). This principle mainly applies to man-made sources of radiation. Natural background radiation levels tend to be relatively low, and they are difficult to reduce. Radiation exposures are additive, regardless of source, and therefore no increases should be taken lightly.

Hofmann et al. (35) and Forastiere et al. (36) conducted epidemiological studies in areas in China and Italy, respectively, where background doses are naturally about twofold higher than in surrounding regions. Although trends were present, neither study was able to statistically prove a relationship between the elevated doses and mortality or lung cancer occurrences in man. Thus, it is probably unrealistic to attempt to quantify health effects due to changes in background dose as low as those predicted by Okamoto as a result of acid rain (1).

Simulations

Approach to Modeling

We sequentially used three separate models to investigate the effects of acid rain on radiological dose to man. The first was an equilibrium water chemistry model (SOLMNQ) documented by Goodwin and Munday (37). We used it to examine the effects of pH changes on Ra and Cs speciation in porewater from a sandy, acidic Canadian Shield soil. The results showed that normal concentrations of Ra and Cs in soils are too low for precipitation. In general, the speciation of solution-phase Ra and Cs was simple, making a partition coefficient or K_d model appropriate to simulate effects of pH on Ra and Cs mobility. Our literature review gave general guidance to estimate the dependence of K_d on pH and we bracketed these estimates to include a range of simulation cases.

The second model was a detailed, unsaturated soil, nuclide transport model (SCEMR). This validated model, featuring atmospherically driven soil water flow and a capability for long-term predictions (38), is described in more detail in the next section.

The final model was a dose calculation model (LIMCAL) documented by Zach (39). It computes a radiation dose to a member of a critical group that derives all the food and water from the contaminated source given steady-state radionuclide concentrations in soil and surface water. It includes algorithms to consider only calorie-consistent dietary intakes.

Soil Radionuclide Release and Migration Modeling

Solute Transport Model Application. SCEMR, developed to predict the transport and redistribution of radionuclides in the unsaturated soil zone, was used to predict the long-term behavior of Ra and Cs in surface soil. The ability of this model to predict solute transport and plant uptake has been validated in previous studies (38–40). This model computes water and solute movement in a five-layered soil profile. Water flow through the soil is computed using an iterative solution of the Darcy equation and depends upon the moisture content profile, the water absorption by roots, and the soil hydraulic properties. Supply and loss of moisture at the soil surface is computed from daily energy balance calculations and thus responds to seasonal trends. Long-term estimates of up to 200 years rely on repeated use of meteorological parameters measured throughout recent years. Solute movement is retarded based on K_d values specific to each element and soil horizon. To simulate acid rain conditions we altered the retardation of Ra and Cs by adjusting the K_d values in SCEMR in accordance with known changes in soil pH. The soil physical and chemical properties as well as the initial Ra and Cs soil concentrations were selected to represent soils most sensitive to acid rain resulting from low natural buffering capacity.

Model Inputs. SCEMR inputs include daily climatic data (rainfall, solar radiation, temperature, wind speed, vapor pressure), soil microhydrologic properties for several soil water layers (bulk density, field capacity, porosity), and soil chemical properties for several soil chemistry layers (cation exchange capacity, nuclide K_d value). We specified characteristics for the five layers to represent a 56.5-cm deep Brunisol soil with a thin, 1.5-cm deep litter layer, a 10-cm thick Ae horizon and a B horizon with three layers, 8-, 8-, and 29-cm thick, respectively. The soil chemistry parameters were varied with each of the five layers. This Brunisol soil had been previously used in experimental work, and its physical and chemical properties have been reported elsewhere (41). Rainfall and the other climatic data were obtained from a standard weather station at Geraldton, Ontario, Canada (Table 2). Geraldton was selected because it had the long-term data required to run SCEMR, and the data are typical of the central Ontario portion of the Canadian Shield in which acid rain is of much concern.

Determination of Soil Chemical Properties and Other Model Inputs. Initial soil concentrations for Ra were taken from Sheppard (14) and initial soil concentrations for Cs from Gillham et al. (22), as shown in Table 3. Other model inputs were directly derived from earlier experiments or from the literature and are given in detail by Sheppard et al. (42). Initial model soil-moisture contents in all layers were set at 90% of field capacity. Radioactive decay is not modeled in SCEMR. The radiological half-life of ^{226}Ra

Table 2. Monthly summaries of meteorological parameters for Geraldton, Ontario.

Parameter	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
						Month						
Total precipitation, cm	5.0	1.3	4.1	7.4	8.1	8.4	10.7	6.5	7.4	8.0	8.8	2.0
Mean daily solar radiation, MJ/m²	5.0	9.3	13.4	15.8	15.5	16.8	19.5	15.3	9.8	7.0	3.9	3.1
Mean daily vapor pressure, Pa	180	220	350	640	990	1670	2190	1870	1120	790	580	360
Mean daily temperature, °C	−15	−10	−5	4	11	20	23	20	11	6	0	−6
Mean daily wind speed, km/hr	10	10	12	15	12	15	13	14	14	14	13	10

is 1600 years and that of ^{137}Cs is 30.17 years. Omitting decay for ^{137}Cs tends to increase dose estimates for man.

Dependence of Mobility on pH. Our review of the literature did not reveal a unique relationship between pH and mobility of Ra or Cs. Instead, a range of effects was indicated. Our modeling approach was to select a measured profile of K_d values to use as a standard, and then to modify this profile through a reasonable range of values corresponding to acid rain conditions. For Ra, we chose a profile of K_d values based on the means reported by Sheppard et al. (*43*) and a range of solubility through the profile derived from Rusanova (*19*). For Cs, we chose a profile of K_d values measured in the Brunisol soil (*41*). All of these measurements of K_d were based on data from samples collected in the field and reflect stable equilibria. The values used for the standard K_d profiles are listed in Table 3.

We modified the K_d values in the top two or three layers of the soil to reflect the effects of acid rain. We reduced the surface-layer K_d by factors of 0.5, 0.2, and 0.1 and modified the values for the second and third layers to create smooth profiles. We also included a profile of increased K_d values (Table 3) to fully demonstrate the response of the simulations to changes in K_d. Based on our literature survey, we expected that acid rain could cause a shift from the standard profile to a situation resembling profiles 2 or 3 (Table 3).

Table 3. K_d values for all simulation cases by soil layer for Ra and Cs and the initial concentrations.

Soil layer	K_d profile cases, mL/g					Initial soil concentration, µg/g
	1[a]	2	3	4	5	
Ra						
Litter	1000	500	200	100	2000	1.2×10^{-6}
Ae	500	250	100	100	1000	1.2×10^{-7}
B	200	100	100	100	400	1.2×10^{-7}
B	100	100	100	100	100	1.2×10^{-7}
B	100	100	100	100	100	1.2×10^{-7}
Cs						
Litter	43.8	20	8	4	80	1595
Ae	20.6	10	4	2	40	737
B	17.2	8	3	2	17	81
B	2.0	2	2	2	2	81
B	2.0	2	2	2	2	81

[a]Standard K_d profile based on estimates from the literature (see text), profiles 2 to 5 are modifications of the standard profile to bracket possible effects of acid rain.

Results of the Simulations

Soil Concentration Changes and Predicted Root Uptake of Ra and Cs. The changes estimated by SCEMR in the total soil Ra concentration in the surface layer (Fig. 3) were almost linear with time. Radium was fairly immobile, and initial concentrations were very low. The standard K_d profile varied by an order of magnitude (Table 3). This profile yielded a relatively simple response with time and changes in mobility. There was a continuous loss of Ra from the soil with an approximate residence half-time (assuming a first-order rate of loss) of about 4000 years. This profile reflects natural weathering processes, and the half-time of 4000 years seems reasonable. Decreases in K_d, to simulate effects of acid rain (profiles 2, 3 and 4 in Table 3), decreased the residence half-time. Reduction in K_d by factors of 0.5, 0.2, and 0.1 lead to reductions of half-time to 2000, 800, and 300 years, respectively. Therefore, a change in K_d of less than an order of magnitude results in a proportionate change in the release of Ra. This confirms one of Okamoto's assumptions.

To avoid having to arbitrarily select a type, size, and culture system for the plants, we have not simulated plant shoot concentrations. The cumulative uptake of Ra by roots is also almost linear with time (Fig. 4). The lines for the various profiles differ by constant factors throughout the simulation. If the results of the standard K_d profile are taken as 100%, the results of K_d profiles 2, 3, and 4 changed 200, 500, and 600%, respectively. As with the soil concentrations, changes in K_d of less than an order of magnitude resulted in proportional changes in plant uptake.

The estimated changes in the total soil Cs concentration in the soil surface layer (Fig. 5) were not as simple as for Ra. Cs was quite mobile and initial concentrations were fairly high. The standard K_d profile varied by less than an order of magnitude (Table 3). Under these conditions, the surface layer accumulated Cs for another 120 years. At lower K_d values, the peak occurred earlier, at 60, 30, and less than 20 years for K_d values 0.5, 0.2, and 0.1 (profiles 2, 3, and 4 in Table 3) of the standard profile. Associated with the earlier peaks are higher peak concentrations. Fallout ^{137}Cs is decreasing with time in soils in Canada mainly due to radioactive decay. It may be that soils have not reached a steady state with respect to Cs loss, as suggested by the model. After the Cs concentrations
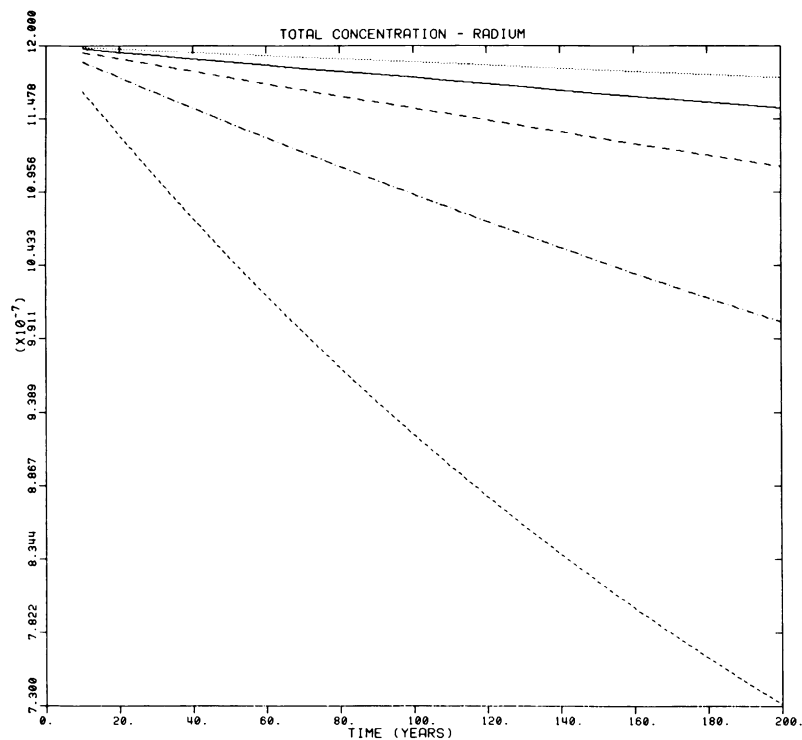
FIGURE 3. Concentration of Ra in the surface layer of soil estimated over 200 years. The lines represent K_d profiles described in Table 3 as 1 (——),
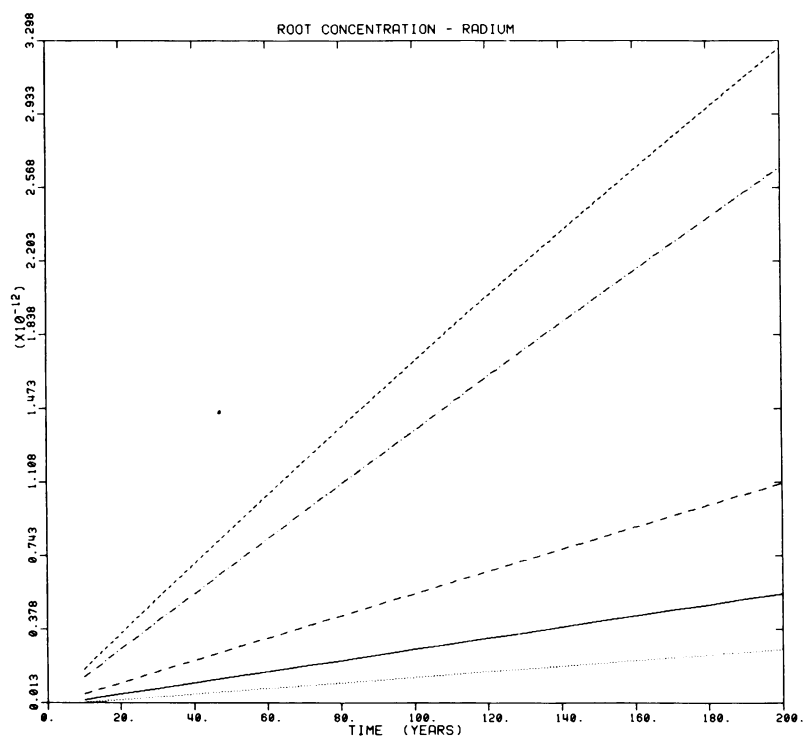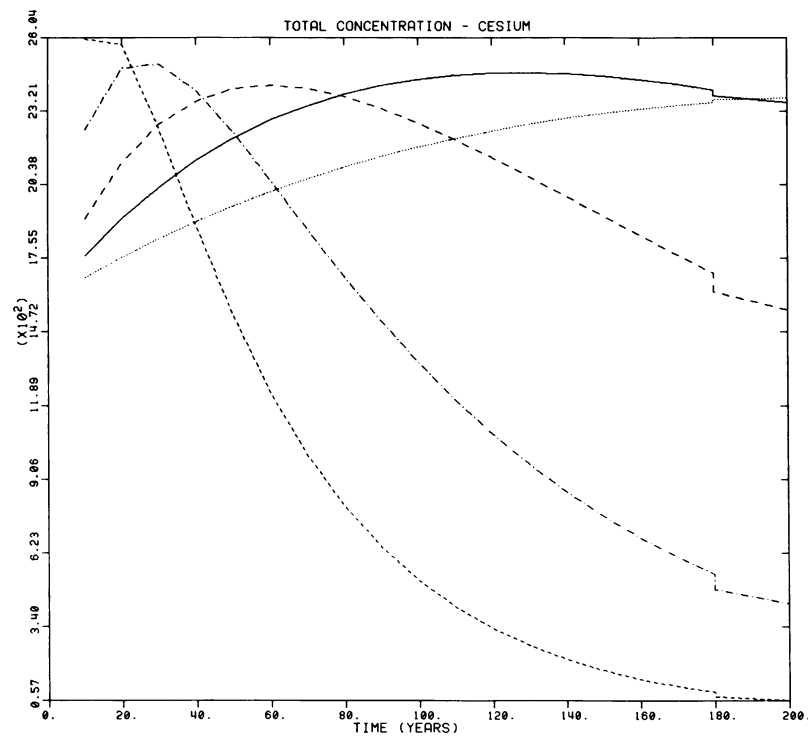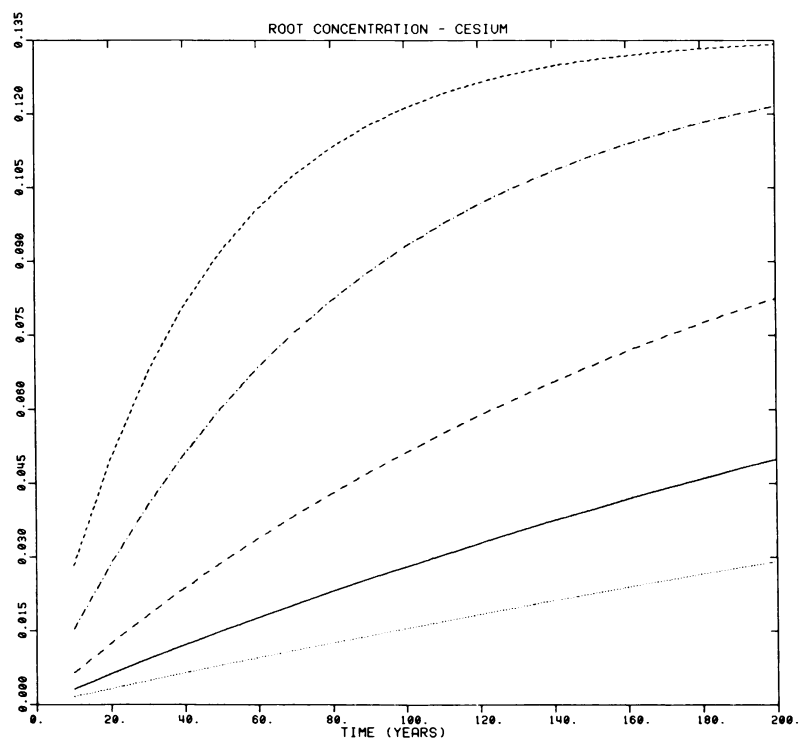2 (— — —), 3 (—·—·), 4 (- - - - -), and 5 (· · · · · ·).

FIGURE 4. Cumulative uptake of Ra by roots estimated over 200 years. The lines represent K_d profiles described in Table 3 as 1 (——),
2 (— — —), 3 (—·—·), 4 (- - - - -), and 5 (· · · · · ·).

FIGURE 5. Concentrations of Cs in the surface layer of soil estimated over 200 years. The lines represent K_d profiles described in Table 3 as 1 (———),
2 (— — —), 3 (—·—·—), 4 (- - - - -), and 5 (·······).

FIGURE 6. Cumulative uptake of Cs by roots estimated over 200 years. The lines represent K_d profiles described in Table 3 as 1 (———),
2 (— — —), 3 (—·—·—), 4 (- - - - -), and 5 (·······).

reached their maxima, they decreased and ultimately approximated a first-order rate of loss. This rate of loss would be steeper had we incorporated radioactive decay. The retention half-times were about 200, 60, and 32 years for K_d profiles 2, 3, and 4, respectively.

The root uptake of Cs is more complex (Fig. 6) than that for Ra. However, the effect of changes in K_d were similar. After 100 years and expressed as a percent of the standard profile, the root uptake was 186, 330, and 430% for profiles 2, 3, and 4, respectively. These values reflect the higher peak soil concentrations that occurred at lower K_d values. The relationship between changes in K_d and changes in root concentration are, therefore, not exactly proportional.

Radiation-Dose Consequences as a Result of Acid Rain. The results of the simulations for Ra and Cs show a more complex relationship between mobility and impact on man through plant uptake than the approximation used by Okamoto (*1*). However, when the K_d, our indicator of mobility, varies by a factor of two, the resulting impact also varies by a factor of about two, as indicated by plant uptake. Thus, the assumption that mobility and impact are directly proportional may be valid within the range of effects on mobility resulting from acid rain.

We used estimates of concentrations of radionuclides in plants to compute doses to man. Models such as LIMCAL compute these doses, summarizing the contributions from various sources of food and water. We modeled uptake by a generic plant, as opposed to computing a range of values for a variety of plants. A twofold change in plant concentration resulted in a twofold change in dose to man.

Conclusions

Natural and anthropogenic radionuclides, especially ^{226}Ra and ^{137}Cs, are among those soil materials that become more mobile due to acid rain. Generally, a decrease in soil pH by 1 unit will cause increases in mobility and plant uptake by factors of 2 to 7. Thus, Okamoto's dose estimate may be too low.

Our model predictions for acid rain conditions showed essentially direct proportionality between the mobility of the nuclides and the dose to man. We conclude that a decrease in pH of 1 unit may increase the mobility of Ra and Cs by a factor of 2 or more. This will lead to increases in plant uptake and a similar increase in dose to man. The literature on acidification related to radionuclide mobility is very scant. Further research, particularly in a fieldlike setting, is clearly required to more effectively quantify the dose consequences of acid rain.

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